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This project encompassed experimental and theoretical research aimed at understanding the underlying physics of dynamically driven magnetization reversal in magnetic nanoparticles. Nanoparticle arrays comprising circular, elliptical and cylindrical dots were patterned by e-beam lithography. The samples were characterized by AFM, MFM and dc magnetization measurements, and their dynamic properties determined by ferromagnetic resonance (FMR) measurements. Detailed numerical solutions of the micromagnetic equations of motion have explored the origin of the peaks observed in the FMR spectrum of the ferromagnetic arrays.

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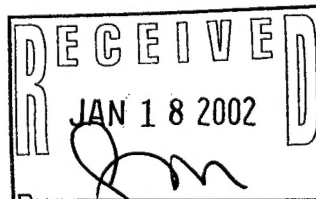
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Sincerely,

Venkat Chandrasekhar

Coherent Dynamics of Magnetic Particles

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Final Progress Report

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Collaborations

Peter Schiffer, Penn State University

Vitali Metlushko, University of Chicago

Lance DeLong, University of Kentucky

Summary

This project encompassed experimental and theoretical research aimed at understanding the underlying physics of dynamically driven magnetization reversal in magnetic nanoparticles. Nanoparticle arrays comprising circular, elliptical and cylindrical dots were patterned by e-beam lithography. The samples were characterized by AFM, MFM and dc magnetization measurements, and their dynamic properties determined by ferromagnetic resonance (FMR) measurements. Detailed numerical solutions of the micromagnetic equations of motion show that the peaks in the FMR spectrum are associated with coupled exchange and dipolar spin-wave modes.

1. Experimental studies of ferromagnetic arrays

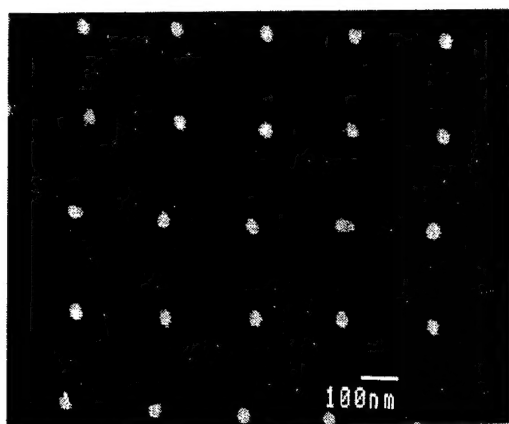


Figure 1 Scanning electron micrograph of an array of Ni dots. Each dot is ~ 80 nm in height, and ~ 25 nm in diameter.

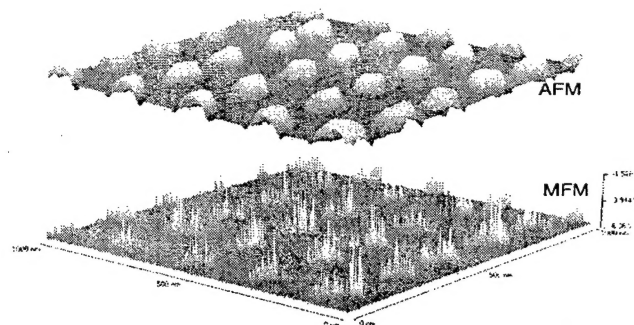


Figure 2 AFM and MFM image of a Ni particle array similar to that shown in Fig. 1.

Fabrication of ferromagnetic arrays

The first part of this project concentrated on the fabrication of ferromagnetic nanoparticle arrays by electron-beam lithography. The fabrication was carried out by post-doc Sukkoo Jung. Our first aim was to fabricate arrays with particle sizes as small as possible. After some initial work, we were able to fabricate Ni arrays with individual particle of height ~ 80 nm and diameter 25 nm. Figure 1 shows a scanning electron micrograph of one such array.

The particle arrays were characterized by MFM, dc magnetization measurements (done in collaboration with Peter Schiffer, now at Penn State) and FMR measurements. Figure 2 shows an example of a combination AFM/MFM measurement of an array similar to the one shown in Fig. 1. The MFM shows that the particles are single domain, with the magnetization confined to one end of each particle. The magnetic behavior can also be seen in the magnetization curves, which show a clear hysteresis loop (Fig. 3).

However, a combination of MFM, dc magnetization and FMR measurements showed that particles in this size regime were not consistently magnetic, i.e., only some arrays showed magnetic behavior. Only particles with diameters above 100 nm were found to be consistently magnetic. Consequently, in our

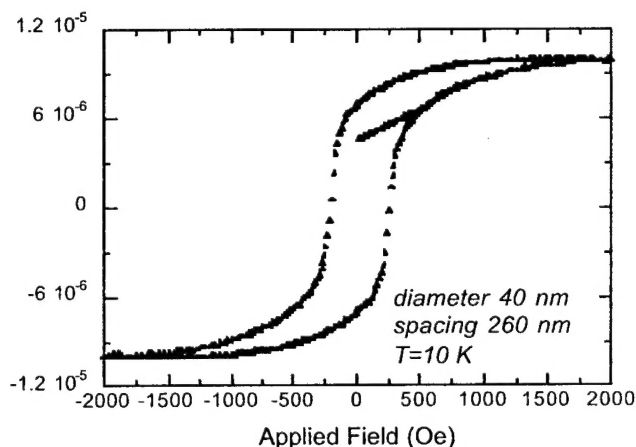


Figure 3 dc magnetization curves of an array of Ni particles of diameter 40 nm and spacing 260 nm, taken with a SQUID magnetometer by Joseph Snyder and Peter Schiffer, Penn State University.

subsequent experiments, we fabricated arrays of particles with elliptical and circular shapes, with all lateral particle dimensions > 100 nm. In addition, we also switched to using permalloy rather than Ni as the ferromagnetic element, since permalloy has a narrower intrinsic linewidth for FMR studies. Figure 4 shows a SEM of one such array of circular elements fabricated by e-beam lithography, with circular nanoparticles of diameter 500 nm separated by 100 nm from their neighbors. Each particle is 70 nm in height.

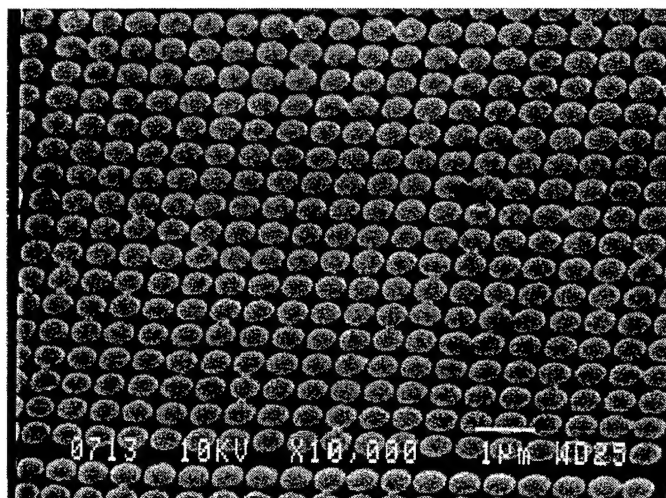


Figure 4 Scanning electron micrograph of an array of circular permalloy disks fabricated by e-beam lithography at Northwestern. The diameter of the disks is 500 nm, their thickness 70 nm, and the center-to-center distance between disks is 600 nm.

Low amplitude FMR

After fabrication and characterization of the ferromagnetic particle arrays, the FMR spectrum of the arrays was measured. The arrays show a number of new and interesting features in their FMR spectra.¹ As an example, we show in Fig. 5 differential FMR spectra for a square array of circular permalloy particles of diameter 530 nm and height 85 nm, with a lattice spacing of 600 nm. This sample was fabricated by e-beam lithography, and measured in a Varian spectrometer at room temperature, in the EPR facility at Northwestern. The dc magnetic field is applied in the plane of the array, and the frequency of the ac field is 9.37 GHz. Five spectra are shown, each corresponding to a different angle of the dc field with respect to the array: 0° corresponds to the field being aligned along one of the principal axes of the array. In all the spectra, a large peak is seen at ~ 1000 Oe, corresponding to the uniform precession mode. At lower fields, a number of additional peaks are observed. In the classical analysis of Kittel, these would correspond to exchange spin-wave modes. In this sample, the number and position of these peaks depends on the angle between the dc magnetic field and the axes of the square lattice. As each permalloy particle here has circular symmetry, the only thing that could be changing as a function of this angle is the dipolar coupling between the particles. This implies that these peaks correspond to spin-waves which have at least some dipolar contribution to their energy.

Even more pronounced is the behavior of the resonance peaks at fields above the uniform resonance mode. When the magnetic field is aligned along the diagonal of the square lattice, a number of peaks are observed which are almost completely absent when the field is aligned along one of the principal axes. (See, for example, the peak at ~ 1000 Oe on the $=45^\circ$ curve.) This shows that a large contribution to the energies of these modes comes from dipolar interactions. For comparison, Fig. 4 shows similar FMR spectra for an array of elliptical permalloy particles with long axis 500 nm, short axis 250 nm, thickness 85 nm, and a lattice spacing of 1 μ m. Here, 0° corresponds to the external dc magnetic field being aligned along the major axis of the ellipse. A broad peak is observed at ~ 600 Oe. This corresponds to the uniform mode peak. As the magnetic field is rotated, this resonance moves up in field, until it is out of the field range of the spectrometer when the field is aligned along the short axis of the ellipse. This shift is associated with demagnetization effects due to the geometry of each particle. The surprising feature, however, is the absence of even a hint of satellite peaks on either side of the uniform mode peak. This is in

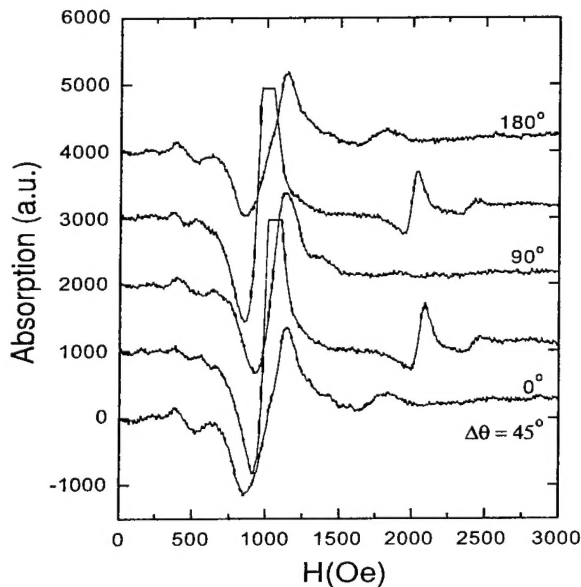


Figure 5 Room temperature FMR spectra of a square array of circular permalloy disks similar to that shown in Fig. 1, as a function of the angle between the dc magnetic field and a principal axis of the square array. The diameter of each disk was 530 nm, thickness 85 nm, and the array lattice spacing was 600 nm. The peak for the uniform mode for the curves corresponding to 45° and 90° were limited by the gain of the spectrometer.

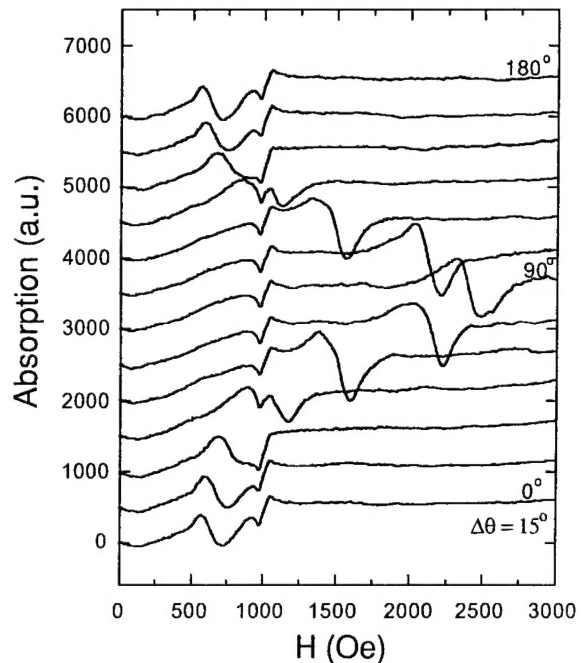


Figure 6 Room temperature FMR spectra for a square array of elliptical permalloy particles, as a function of the angle θ between the dc magnetic field and the major axis of the ellipse. Each ellipse has a long axis of length 500 nm, short axis of length 250 nm, and the lattice has a spacing of 1 μm . The small peak at $H \sim 1000$ Oe whose position does not shift with angle is associated with a residual film on the sample substrate.

marked contrast to the circle arrays, and indicates that it is more difficult to excite spin-wave modes in the elliptical geometry, where the shape anisotropy aligns the magnetization along the major axis of the ellipse. Similar behavior is also observed for antidot arrays, i.e., films with an array of holes patterned by electron-beam lithography. These arrays were patterned by Vitali Metlushko at Argonne National Laboratory. These arrays also show a 4-fold symmetry of the FMR spectrum under certain conditions.

The ultimate goal of our experiments is to determine the conditions that would allow reversing the magnetization of a single ferromagnetic particle by the application of a high-frequency pulse. It is clear that a knowledge of the spin-mode structure is essential to explore the possibility of using pulsed microwave radiation to reverse the magnetization of a single ferromagnetic nanoparticle.

2. Numerical Studies of Magnetization Dynamics

The earlier theoretical investigations of spin-wave modes have involved looking at the influence of either exchange interactions or dipolar interactions alone in very specific geometries, which gave solutions that could be tackled analytically. For the particle sizes of interest here, both

the dipolar and exchange interactions are of importance. Furthermore, the shapes of the particles do not admit easy analytical solutions, so that numerical solutions of the Landau-Lifshitz equations are required.

For solving the micromagnetic equations of motion, we have used the public domain micromagnetic solver, OOMMF, available from the NIST website.² This program numerically integrates the Landau-Lifshitz equation, taking into account dipolar interactions, exchange interactions, and the detailed geometry of the particle, as well as specific magnetic parameters (such as the crystalline anisotropy) of the magnetic material. We have successfully used this program to model the FMR response of circular ferromagnetic particles.^{1,3} In order to do this, we generate the time evolution of the magnetization under the influence of the dc and fixed frequency microwave field. We then extract the amplitude of the magnetization response at the frequency of the microwave field. This is done over a range of values of the dc magnetic field. Figure 7(a) shows the numerically

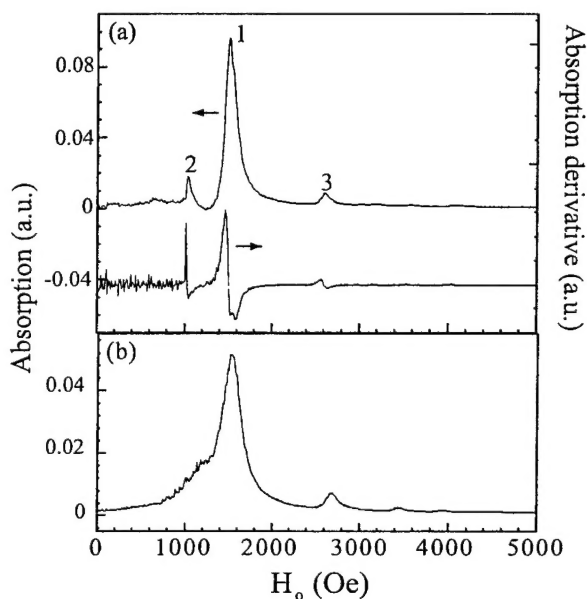


Figure 7 . (a) Calculated FMR absorption spectrum for a 0.5 μm diameter permalloy disk. The lower curve is the derivative of the absorption curve. The saturation magnetization is $M_S=8.47 \times 10^5$ A/m, exchange stiffness $A=1.3 \times 10^{-11}$ J/m, and damping constant $\alpha=0.05$. (b) Identical to (a), except $A=0$.

calculated FMR spectrum of a 0.5 μm diameter permalloy disk, taking into account both exchange and dipolar contributions. As can be seen, the resulting spectrum is qualitatively very similar to what we observe experimentally. Figure 7(b) shows the spectrum calculated with parameters identical to those of Figure 7(a), except with the exchange constant set to 0. The peak below the uniform mode peak (labeled '1' in Figure 7(a)) has essentially disappeared, showing that the primary contribution to the energy of the corresponding spin-wave mode came from exchange interactions. The positions of the peak above the uniform mode peak (for example, the one labeled '3' in Figure 7(a)), are also shifted slightly (by a few hundred Oe, which is difficult to see in the scale of the plot), demonstrating that only a small fraction of the energies of the corresponding spin-wave modes also come from exchange interactions (most of the energy comes from dipolar interactions). By varying the parameters in the numerical simulations to fit the experimental data, one can determine the relevant microscopic magnetic parameters of the samples.

In addition to the ability to calculate the FMR spectra, the OOMMF program allows one to visualize directly the nature of the modes by plotting the time evolution of the magnetization under the influence of the ac field. Figure 8 shows the time evolution of the magnetization distribution through one cycle of the applied ac field for the three peaks marked '1,' '2,' and '3' in Fig. 7(a).

In Fig. 8(a), which shows the time evolution for the peak '1,' almost all of the magnetic moments move in phase, demonstrating clearly that this resonance indeed corresponds to the uniform mode peak. Figure 8(b) shows the time evolution for the peak '2.' In this case, the static distribution of moments is distinctly inhomogeneous, with small oscillations about this inhomogeneous state in

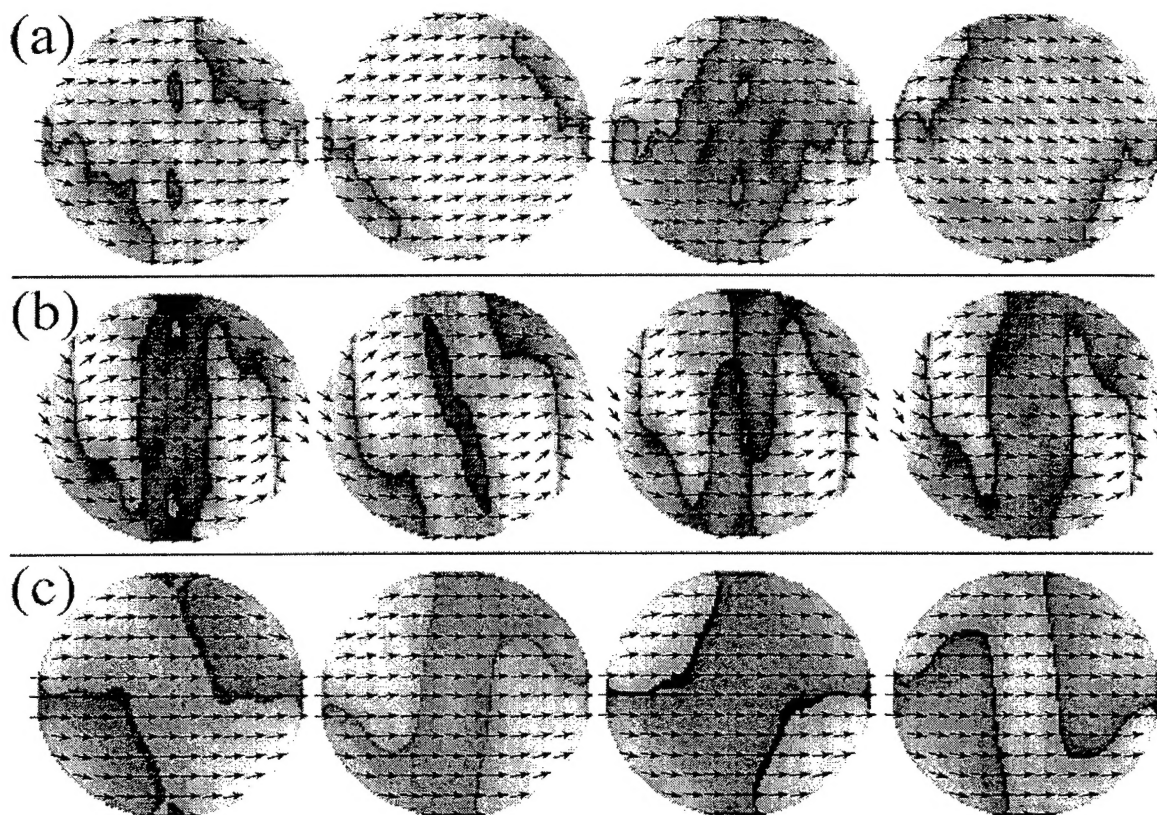


Figure 8 Each panel represents the time evolution of the magnetization distribution through one cycle of the ac field (at phases $\sim 0, \pi/2, \pi, 3\pi/2$) for a 0.5 μm diameter permalloy particle at magnetic fields corresponding to the peaks labeled 1-3 in Fig. 7(a). (a) peak '1'; (b) peak '2'; (c) peak 3. H_{ext} is aligned along the x -axis, and the grayscale plot denotes the angle between the y component of M and the x -axis.

response to the ac field. Finally, Fig. 8(c) shows the time evolution for the spin-wave mode corresponding to peak '3.' This corresponds to a spin-wave mode which is primarily dipolar in nature. Here, the magnetic field is strong enough so that the most of the magnetic moments are essentially aligned along the direction of the external magnetic field. Although this example involved a sinusoidal ac field, the program allows us to simulate any waveform, including a 'chirped' waveform, which will be essential in the latter part of the project. Our experience with the program so far has been very positive, and we believe it will help us rapidly focus on the appropriate strategies for achieving our experimental goals.

References

1. *Ferromagnetic resonance in periodic particle arrays*, S. Jung, B. Watkins, L. DeLong, J.B. Ketterson and V. Chandrasekhar, submitted to Physical Review Letters.

2. M.J. Donahue and D.G. Porter, URL: <http://math.nist.gov/oommf>.
3. *Numerical simulations of the FMR response of magnetic nanoparticles*, S. Jung, J.B. Ketterson and V. Chandrasekhar, in preparation.

3. Publications and Presentations

Presentations

1. *Ferromagnetic Resonance of Permalloy Films with Patterned Array of Holes*, Byron Watkins, Jeff Feller, J. B. Ketterson, Lance DeLong, Vitali Metlushko, Jung Sukkoo, Venkat Chandrasekhar, APS March Meeting, March 12-16, 2001, Seattle, Washington.
2. *Ferromagnetic resonance response of electron-beam patterned arrays of ferromagnetic nanoparticles*, Sukkoo Jung, Byron Watkins, Jeffrey Feller, John Ketterson, Venkat Chandrasekhar, APS March Meeting, March 12-16, 2001, Seattle, Washington.

Publications

1. *Ferromagnetic resonance in periodic particle arrays*, S. Jung, B. Watkins, L. DeLong, J.B. Ketterson and V. Chandrasekhar, submitted to Physical Review Letters.
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